

**REMARKS**

Claim 1 is amended in order to more particularly point out, and distinctly claim the subject matter which the Applicants regard as their invention. The Applicants respectfully submit that no new matter has been added. Claim 29 is cancelled without prejudice or disclaimer. It is believed that this Amendment is fully responsive to the Office Action dated February 4, 2009.

In the Office Action, Claims 1-6 and 8 were rejected under 35 U.S.C. §102(b) as being anticipated by Takada et al. in their publication in the Journal of Sol-Gel Science and Technology, titled "Control of Particle Size Distribution of CdS quantum Dots in Gel Matrix"; and Claim 7 was rejected under 35 U.S.C. §103(a) as being unpatentable over Takada et al. in their publication in the Journal of Sol-Gel Science and Technology, titled "Control of Particle Size Distribution of CdS quantum Dots in Gel Matrix" as applied to Claim 1 above, and further in view of Chia et al. in their publication in SPIE, titled "Cadmium Telluride quantum dot-doped glass by the sol-gel technique. Reconsideration and removal of these rejections are respectfully requested in view of the amendment to Claim 1 and the following remarks.

Organic compounds are decomposed by heating.

In our previous response, we argued, based on the disclosure in C.J. Brinker et al., J. Non-Cryst. Solids, 72, 345 (1985), that when heated, an organic compound is decomposed and desorbed.

However, the Examiner states that when a semiconductor superfine particle-dispersed glass is heated, only water is desorbed (see page 9, second line from the bottom to page 10, line 8 of the Office Action).

Regarding Brinker, it is respectfully submitted that the Examiner's above-stated comments are incorrect. The Examiner's opinion may be based on the description on page 351 of Brinker, 13th line from the bottom to 4th line from the bottom. The Examiner's incorrect view that only the removal of water causes weight loss may be from reading only the first three lines of this portion.

In reality, however, this portion describes the following:

"Weight loss in this region is attributed both to the removal of water ... and to the oxidation of carbonaceous residues ...." More specifically, there are two factors that cause weight loss: the "removal of water", and the "oxidation of carbonaceous residues". As is clear from the above, two factors, i.e., the "removal of water" and the "oxidation of carbonaceous residues" cause weight loss. The "oxidation of carbonaceous residues" indicates that the organic compound is decomposed by heating.

To reinforce this argument, the document A.B. Wojcik, et al., J. Sol-gel Sci. Tech., 5, 77 (1995) is cited, which also describes decomposition of an organic compound by heating. This document is referred to more clearly and quantitatively explain the decomposition. Enclose, as notes, are the present inventors' comments on three portions of the description on page 79 of Wojcik.

Regarding Wojcik, Wojcik describes producing samples by adding a trifunctional alkoxide

(VTES, having a vinyl group) and an organic polymer (HEMA, HDDA, GPTA) to a tetrafunctional alkoxide (TEOS), as used in Brinker, and then solidifying the mixture by a sol-gel reaction. Wojcik further describes heating the samples in air and measuring the weight loss as a temperature function. Wojcik shows the compositions of the samples in Table 1, which is copied below.

*Table 1.* Compositions of the inorganic/organic sol-gel copolymers and their thermal behavior.

Sample code	Composition mol : mol : mol	Silica content		Decomposition onset T (°C)	Weight loss (drying, %)
		Pyrolysis (wt.%)	TGA (wt.%)		
	HEMA : VTES : TEOS				
HEMA 1(#1)	1.67 : 1 : 1.89	68.9	78.1	278	19.0
HEMA 2(#2)	0.9 : 1 : 1.57	66.7	76.2	270	20.7
HEMA 3(#3)	2 : 1 : 1.89	44.4	34.5	230	11.3
HEMA 4(#4)	3 : 1 : 1.72	31.4	27.7	220	9.8
	HDDA : VTES : TEOS				
HDDA 1(#5)	0.56 : 1 : 1.89	83.4	83.2	272	7.5
HDDA 2(#6)	0.8 : 1 : 1.48	52.3	56.7	285	7.9
HDDA 4(#7)	1.11 : 1 : 1.31	38.7	39.3	336	5.2
HDDA 3(#8)	1.11 : 1 : 1.89	32.4	34.1	340	5.1
	GPTA : VTES : TEOS				
GPTA 1(#9)	0.1 : 1 : 2.1	68.2	68.2	345	4.0
GPTA 2(#10)	0.15 : 1 : 0.87	56.7	61.0	350	6.1
GPTA 3(#11)	0.40 : 1 : 2	28.2	28.5	350	7.0
Silica(#12)	100% TEOS	90.3	90.0	—	9.2

Table 1 of Wojcik shows that all the samples #1 to #11 decompose at a temperature of about 220°C to about 350°C. This is also clear from Figs. 5 to 7 of Wojcik. Such weight loss is caused by decomposition of the organic compounds.

A sample consisting of 100% of TEOS is presented as #12 in Table 1. Fig. 5 of Wojcik shows this sample as Silica, and the results are similar to those shown in Brinker.

Sample #12 is produced using a tetrafunctional alkoxide. Therefore, most of the water and organic compound are decomposed into silica when pretreated for about 1 week. This is also clear from the results in Table 1 of Wojcik, which shows that the silica content of Sample #12 is 90% or more. Because silica has a very high boiling point, i.e., 2,230°C, it is not decomposed by heating at about 600°C. In the curve of Silica shown in Fig. 5 of Wojcik, the weight loss up to about 150°C is mainly caused by desorption of free water and adsorbed water. The subsequent mild weight loss is caused by decomposition of the organic compound.

Therefore, as explained above, a combination of Brinker and Wojcik more clearly shows that when heated at about 220°C to about 350°C in air, an organic compound in a glass produced by a sol-gel reaction is thermally decomposed and desorbed. Furthermore, as stated in the previous Argument, there are many publications that state that organic compounds are decomposed by heating; it is respectfully submitted that decomposition of an organic compound by heating is common technical knowledge among persons skilled in the art.

Regarding surface condition of Takada and Chia, Takada produces CdS superfine particles according to the reaction scheme shown in Fig. 2, which is copied below.

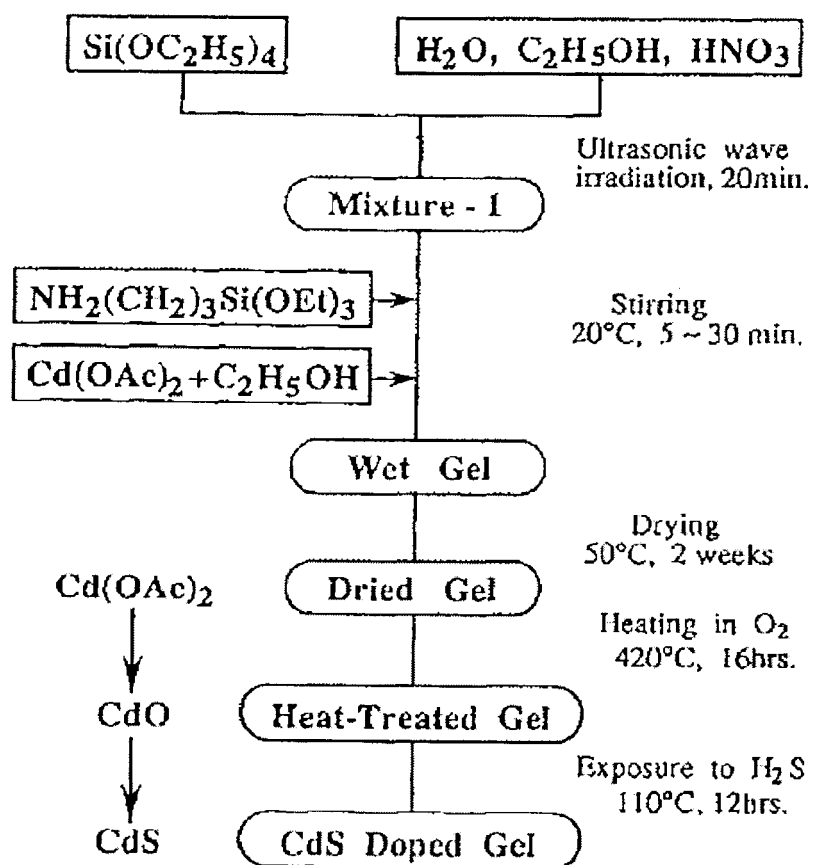


Fig. 2. Procedures used to prepare CdS quantum dots with the controlled particle size by using  $\text{H}_2\text{N}(\text{CH}_2)_3\text{Si}(\text{OC}_2\text{H}_5)_3$  as a bifunctional ligand via the sol-gel process.

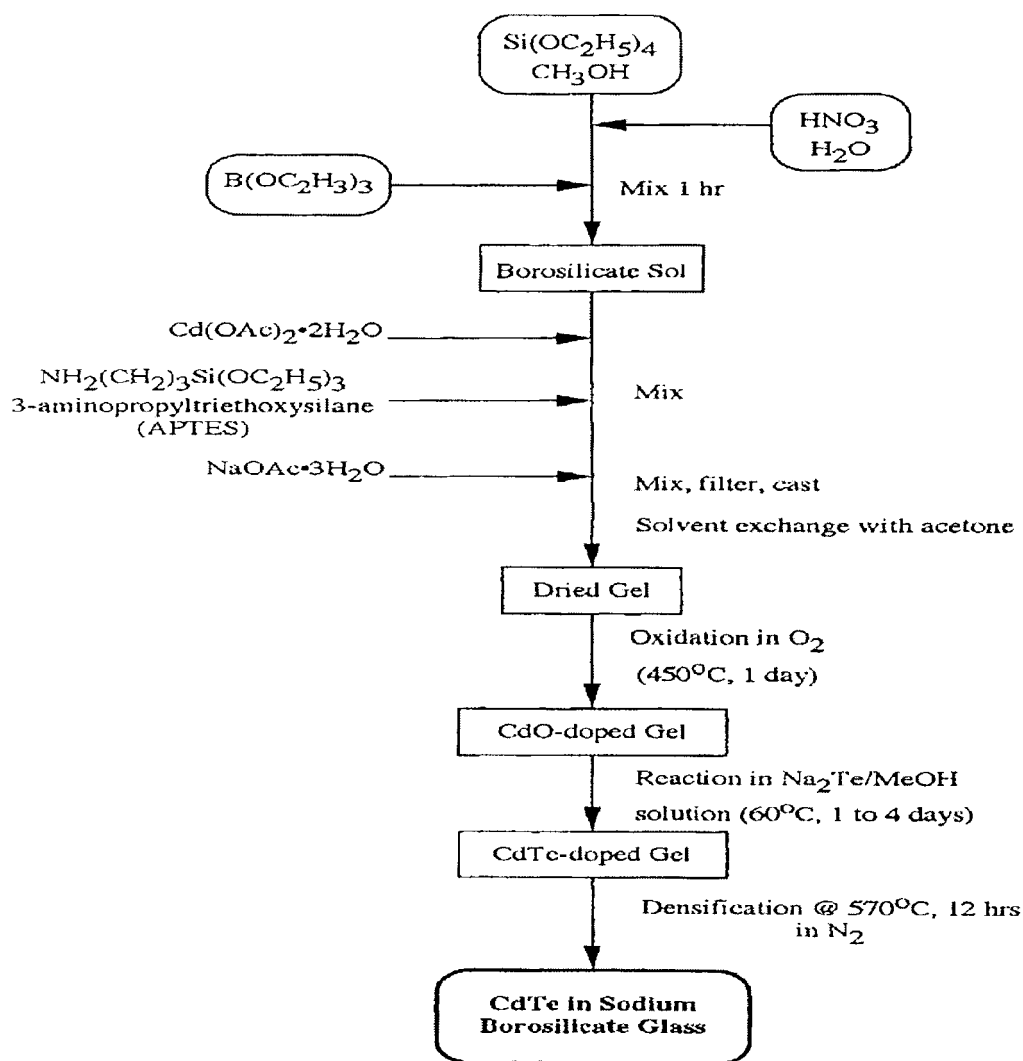
According to Takada, a gel is heated in an atmosphere of oxygen at  $420^\circ\text{C}$  for 16 hours. As explained above, organic compounds thermally decompose at a temperature of about  $220^\circ\text{C}$  to about  $350^\circ\text{C}$ . Thus, organic compounds (such as APS) are almost completely desorbed in Takada. That is, there is nothing that can coat the superfine particles.

Ammonia is decomposed into nitrogen monoxide at about 300°C to about 500°C. Because the amino groups are also considered desorbed from APS in Takada, the superfine particles of Takada are not considered to be protected by APS.

Takada calls the product a "gel". However, as is clear from the above reaction scheme, the final product is a heat-treated gel. The heat-treated gel is a solid having many small pores therein, and the size of the pores is such that molecules can pass therethrough (about 1 nm), and air is present therein. That is, the heat-treated gel is a cluster that appears to be exactly the same as a usual glass. When this cluster is exposed to hydrogen sulfide, the hydrogen sulfide is diffused inside through the small pores, and the O of the CdO superfine particles is substituted by S. However, because the product merely has open pores, it is necessary to remove the semiconductor superfine particles from the glass to coat the particles with a coating agent. Thus, when using the method of Takada, it is physically impossible to disperse semiconductor superfine particles in a glass in a state in which the particles are coated with a surfactant or the like.

Thus, it is respectfully submitted that the semiconductor superfine particles of Takada do not have a coated surface.

Regarding Chia, Chia produces CdTe superfine particles according to the reaction scheme of Fig. 1, which is copied below.



**Fig. 1.** Preparation of CdTe Quantum Dot-doped Sodium Borosilicate Glass by Method (a)

Chia performs heating at  $450^\circ\text{C}$  for one day, as in Takada. Therefore, organic compounds (such as APS) are almost completely desorbed in Chia. That is, there is nothing that can coat superfine particles.

Because ammonia is converted to nitrogen monoxide at about 300°C to about 500°C, the amino groups of APS are presumably desorbed in Chia just as they are in Takada. This also indicates that the superfine particles of Chia are not protected by APS, etc. Therefore, as is clear from the above, the semiconductor superfine particles of Takada and Chia do not have a coated surface. In contrast, according to the present invention, and as recited more clearly in amended Claim 1, "the semiconductor superfine particles have a surface coating of a material other than material found in the silicon-containing solid matrix". That is, the surface of the semiconductor superfine particles is coated. It is respectfully submitted that the present invention is structurally different from Takada and Chia in this point. The present invention is not obvious over Takada or Chia, neither of which describes or suggests coating the surface of the particles.

Regarding the state of the surface of the semiconductor superfine particles of the present invention, as stated above, no surfactants or other substances are present on the surface of the semiconductor superfine particles produced in Takada and Chia. In contrast, according to the present invention, a surfactant, etc. is present on the surface of the obtained semiconductor superfine particles so as to maintain a high fluorescence quantum yield. Regarding this point, the present Specification states the following:

"Examples of such molecules include compounds expressed by the general formula  $\text{HyX1}((\text{CH}_2)_t\text{COOH})_2$  (wherein X1 is N or S, t is an



integer from 5 to 20, and y and z are integers selected so as to satisfy the requirements of the valency of X1), and salts thereof. Specific examples include  $\text{HS}(\text{CH}_2)_t\text{COOH}$ ,  $\text{H}_2\text{N}(\text{CH}_2)_t\text{COOH}$ ,  $\text{HN}((\text{CH}_2)_t\text{COOH})_2$ , and salts thereof. Examples of such salts include sodium, potassium, and other alkali metals salts. It is preferable for t to be an integer from 6 to 15, and more preferably an integer from 7 to 13. Among these, thiols (i.e. X1 is S) are preferable, and mercaptoundecanoic acid ( $\text{HS}(\text{CH}_2)_{10}\text{COOH}$ ) is particularly preferable" (page 11, lines 1 to 11).

Regarding the conditions of superfine particles that emit bright light, the present Specification further describes the following:

"Sulfur-containing compounds can suitably be used for such surface treatments. Typical examples include thiols and other organic surfactants, as well as zinc sulfide. Since semiconductor superfine particles whose surfaces are thoroughly coated using such compounds are incredibly bright, it has been shown in the latest research that emission from each individual particle can be separately detected and optically resolved" (page 2, lines 29 to 35).

Furthermore, Example 3 of the present Specification describes using thioglycerol as a specific example of the surfactant. As is clear therefrom, it is important in the present invention to protect

semiconductor superfine particles with a surfactant or the like, and maintain the protected state.

Regarding reduction of a fluorescence quantum yield by heating, the semiconductor superfine particles of the present invention have a fluorescence quantum yield of 3% or more. The semiconductor superfine particles are produced by a solution method, and then dispersed in a glass in a state in which the particles are protected with a surfactant or the like. As described above, the organic compound to be used for coating the semiconductor superfine nanoparticles is thermally decomposed by heating; no substances such as surfactants are present on the surface, which would result in a sharply reduced fluorescence quantum yield. It is respectfully submitted that it is thus impossible to achieve a fluorescence quantum yield of 3% or more.

A Declaration under 37 C.F.R.1.132 enclosed herewith clearly demonstrates this matter. In Experiment A of the Declaration, a CdTe superfine particle-dispersed glass was produced in the same manner as in Example 1 of the Specification. The obtained CdTe superfine particles had a fluorescence quantum yield of 7%, and an average particle size of about 4 nm. The concentration of the CdTe superfine particles was  $6 \times 10^{-4}$  mol/l.

In Experiment B of the Declaration, the CdTe superfine particle-dispersed glass obtained in Experiment A was heat-treated at about 200°C. A sharply reduced fluorescence quantum yield, i.e., 0.4%, resulted. Even a heat-treatment at such a comparatively low temperature as 200°C resulted

in a sharply reduced fluorescence quantum yield. Thus, if a heat-treatment is performed at 400°C or higher, the fluorescence emission intensity will be reduced even more, and the semiconductor superfine particles will hardly emit any light.

It is thus clear that the semiconductor superfine particles of Takada and Chia cannot achieve a fluorescence emission intensity of 3% or more.

In contrast, the present invention achieves a fluorescence emission intensity of 3% or more. The present invention is thus clearly different from Takada and Chia in this point. Furthermore, Takada and Chia nowhere specifically describe fluorescent emission intensity. Only semiconductor superfine particles with very low fluorescence emission intensity can be obtained in Takada and Chia due to a heat treatment performed to produce semiconductor superfine particles. More specifically, the fluorescent emission intensity of 3% or more achieved by the present invention cannot be achieved by Takada or Chia.

Regarding differences in the formation mechanism of semiconductor superfine particles, there are two mechanisms for producing semiconductor superfine particles: one is a process comprising growing semiconductor superfine particles in a glass (production process (a)), and the other is a process comprising growing semiconductor superfine particles in a solution (production process (b)). These processes are essentially and greatly different from each other. We will explain the differences below:

Process of growing semiconductor superfine particles in a glass (production process (a))

This production process is used in Takada and Chia. In production process (a), cores of semiconductor superfine particles are formed in a glass, and then the particles grow. According to this process, the matrix has a network structure, and semiconductor superfine particles are too large to move therethrough. Only the constituent elements such as Cd, Te, and S move in the form of atoms or ions, and attach to the surface of the superfine particles, whereby the superfine particles grow.

This is described in detail in A. Ekimov, J. Lumin., 70, 1 (1996) (hereinafter referred to Ekimov). As described below, Fig. 1 of Ekimov shows a relationship between the heating time (plotted as the abscissa; unit: hour) and the average particle size (plotted as the ordinate; unit: angstrom) of CdS superfine particles.

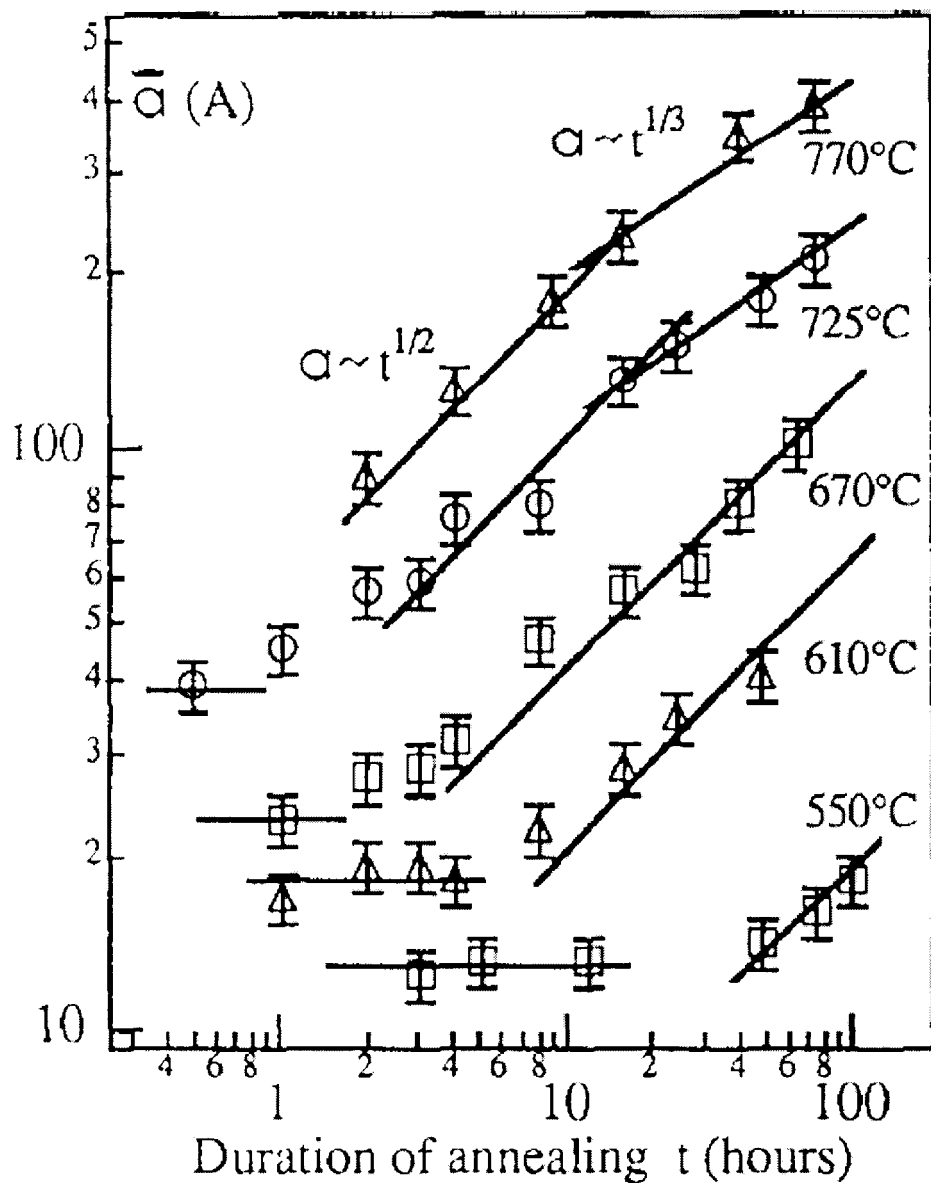


Fig. 1. Dependence of mean radius of CdS nanocrystals in a matrix of oxide glass as a function of annealing duration at various temperatures. Solid lines show slope of the dependencies for different stages of phase decomposition process.

Due to the high viscosity of glass, a heat treatment at a high temperature, i.e., 500°C or higher, as shown in Fig. 1, is necessary to use production process (a). As is clear from the above argument in regarding the Declaration, organic compounds such as surfactants are decomposed when heated at such a high temperature, and thus cannot be used. As stated above, it is physically impossible to coat the surface of semiconductor superfine particles with a surfactant after the particles are grown in a glass.

Therefore, such particles cannot meet the conditions that "the semiconductor superfine particles have a surface coating of a material other than material found in the silicon-containing solid matrix", as recited in amended Claim 1.

Process of growing semiconductor superfine particles in a solution (production process (b))

This process is used in the present invention. In a solution, unlike in a glass, the semiconductor superfine particles themselves can also move. Therefore, unless semiconductor superfine particles are coated with a surfactant or the like, the particles are prone to aggregate. The semiconductor superfine particles of the present invention meet the conditions that "the semiconductor superfine particles have a surface coating of a material other than material found in the silicon-containing solid matrix". Therefore, the particles have no surface defects, and excitons formed by absorption of excitation light are not deactivated, thus resulting in a high fluorescence quantum yield.

As an example of process (b), we cite A. L. Rogach, Mater. Sci. Engin., B69, 435 (2000) (hereinafter referred to as Rogach), which describes the relationship between the growth of CdTe superfine particles in an aqueous solution and in dimethylformamide, absorption, and emission spectrum. Fig. 3 of Rogach shows that when refluxing is performed at a temperature of about 100°C, the semiconductor superfine particles grow from about 2.5 nm to about 5 nm with time, accompanied by the shifting of absorbance and luminescence from the blue to the red side due to quantum size effects.

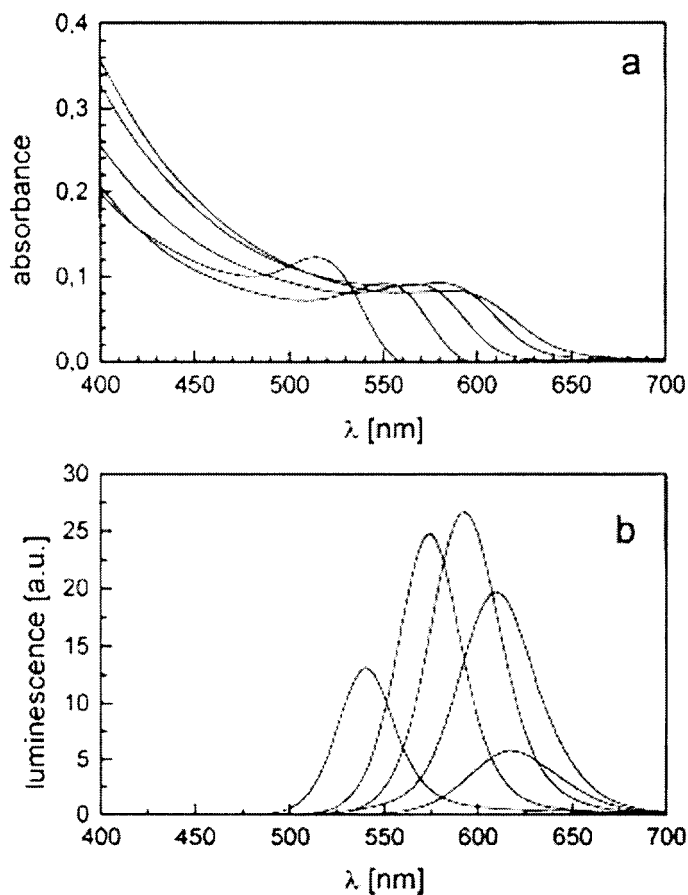


Fig. 3. Optical absorption (a) and luminescence (b) spectra of CdTe nano-crystals (absorption maximum at 515 nm; luminescence maximum at 535 nm; size 2.5 nm) and CdTe(S) nano-crystals (all another spectra: 3–5-nm size range) synthesized in DMF.

In the present invention, the semiconductor superfine particles coated with a surfactant and produced by a solution process are dispersed in a glass by a sol-gel process to stabilize the semiconductor superfine particles. That is, the semiconductor superfine particles are dispersed in a glass in a state in which the particles are coated with a surfactant or the like. Therefore, the particles meet the Claim 1 condition that "the semiconductor superfine particles have a surface



coating of a material other than material found in the silicon-containing solid matrix".

**Additional comments to enable a better understanding of the invention:**

Regarding Claim 29, (cancelled) the Examiner states that the particles are grown within a silica matrix, and thus have a surface coating comprising said matrix. However, it is respectfully submitted that this is not correct. First, the semiconductor superfine particles of the present invention are not produced by growing particles in a silica matrix. According to the present invention, semiconductor superfine particles grown in a solution are dispersed in a glass. This difference results in a structural difference, i.e., whether the particles have a surface coating or not, as explained above.

Regarding fluorescence and wavelength used for nonlinear optical effects, on page 9 of the Office Action, views on the relationship between the fluorescence and wavelength used for nonlinear optical effects are stated. First, the wavelength used for nonlinear optical effects may be in a long wavelength region (a nonresonant region), as stated by the Examiner, or in an absorption wavelength region (a resonance region). These two wavelength types have their own characteristics, and can be selectively used according to the purpose. More specifically, contrary to the Examiner's view, a long wavelength light is not always used.

Seizo Miyata, Nonlinear Optics of Organic Molecules and Polymer, edited by Hari Singh Nalwa, CRC Press, 1977 shows on page 772 that the three-dimensional nonlinear optical constant

(c(3)) increases in an absorption region. However, when absorption occurs, electrons are kept in an excited state for some time (typically on the order of 10 ns, thus usually resulting in defects such as late response. As is also clear from the above, the wavelength of light to be used varies depending on the intended use.

Even when the transition is direct, fluorescence is not always emitted. As stated in our previous Argument, only when surface defects are removed from the surface of the semiconductor superfine particles by using a surfactant or the like, the particles can emit light of a narrow spectral band width with a high fluorescence quantum efficiency at a wavelength near the band gap. This is also clear from the experimental data shown in 3.4. It is respectfully submitted that contrary to the Examiner's views, the band gap is not the only factor that determines whether fluorescence emits.

Regarding nonlinear optical effects, although this strays from the main argument, the Examiner discusses the mode of expression of nonlinear optical effects in paragraph 7 of the Office Action, and states the following: "the nonlinear optical properties stated in Takada et al. have absolutely nothing to do with the light-traveling direction."

However, as is clear from the bolded text ("change in refractive index") in paragraph 7 of the Office Action, NLO properties are based on the refractive index change.

In the previous argument, we explain this matter by using Equation (1):

$$n=n_0+n_2I \quad (1).$$

The refractive index change may be expressed in various ways. As stated by the Examiner, refractive index changes may be expressed as changes in optical bistability, self-phase modulation, and/or phase conjugation. However, refractive index changes may also be expressed as changes in the light-traveling direction. A well-known example of changes in the light-traveling direction is that of light traveling in air bending as it enters water. Specifically, the nonlinear optical properties described in Takada et al. are related to the light-traveling direction.

In view of the amendment to Claim 1, and the above remarks, removal of this rejection is respectfully requested.

In view of the aforementioned amendments and accompanying remarks, Claims 1-8, as amended, are believed to be patentable and in condition for allowance, which action, at an early date, is requested.

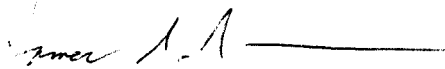
If, for any reason, it is felt that this application is not now in condition for allowance, the Examiner is requested to contact the Applicants' undersigned agent at the telephone number indicated below to arrange for an interview to expedite the disposition of this case.

U.S. Patent Application Serial No. **10/518,216**  
Response to OA dated February 4, 2009

In the event that this paper is not timely filed, the Applicants respectfully petition for an appropriate extension of time. Please charge any fees for such an extension of time and any other fees which may be due with respect to this paper, to Deposit Account No. 01-2340.

Respectfully submitted,

KRATZ, QUINTOS & HANSON, LLP

  
James N. Baker  
Agent for Applicant  
Reg. No. 40,899

JNB/ak

Atty. Docket No. **080308**  
Suite 400  
1420 K Street, N.W.  
Washington, D.C. 20005  
(202) 659-2930



**23850**

PATENT & TRADEMARK OFFICE

Enclosure:    Petition for Extension of Time  
                  Information Disclosure Statement  
                  Declaration under 35 C.F.R. §1.132